Perturbative evolution of far off-resonance driven two-level systems: Coherent population trapping, localization, and harmonic generation

V. Delgado[1] and J. M. Gomez Llorente[2]

Departamento de Física Fundamental II,
Universidad de La Laguna, 38205-La Laguna, Tenerife, Spain

Abstract

The time evolution of driven two-level systems in the far off-resonance regime is studied analytically. We obtain a general first-order perturbative expression for the time-dependent density operator which is applicable regardless of the coupling strength value. In the strong field regime, our perturbative expansion remains valid even when the far off-resonance condition is not fulfilled. We find that, in the absence of dissipation, driven two-level systems exhibit coherent population trapping in a certain region of parameter space, a property which, in the particular case of a symmetric double-well potential, implies the well-known localization of the system in one of the two wells. Finally, we show how the high-order harmonic generation that this kind of systems display can be obtained as a straightforward application of our formulation.

PACS number(s): 42.50.-p, 42.50.Ct

I. INTRODUCTION

Numerical investigations in driven symmetric bistable systems have revealed a number of striking quantum phenomena [1]. On one hand, Grossmann *et al.* [2] have shown that the parameters of the external driving field can be appropriately tuned so as to produce coherent suppression of tunneling, a property that can be used to localize the quantum system in one of the two wells. By properly choosing the shape of the driving laser pulse, localization can be achieved even if the system is initially in a delocalized eigenstate: the driving field can take it into a localized state and then keep it there [3]. On the other hand, this system has also been shown to exhibit high-order harmonic generation in a region of parameter space which overlaps to a certain extent with that where localization occurs [4]. Harmonic generation is a consequence of the fact that, in a strong field, the induced dipole moment responds with frequencies that are integer multiples of the laser frequency, thus giving rise to the appearance of the corresponding peaks in the emission spectrum.

[1]e-mail: vdelgado@ull.es
[2]e-mail: jmgomez@ull.es

An interesting aspect of driven double-well systems is that many of their relevant features can be captured in a simple two-level model. Using this kind of approach it has been shown [5] that, in a wide parameter range [6], the localization conditions can be correctly obtained as a zeroth-order result of a perturbative analysis in the small parameter $\Delta_0/\omega_{\rm L}$, with Δ_0 being the transition frequency of the two-level system and $\omega_{\rm L}$ the driving field frequency. Specifically, in this far off-resonance regime ($\Delta_0/\omega_{\rm L} \ll 1$), localization was found to occur at the zeros of the Bessel function $J_0\left(2\Omega_0/\omega_{\rm L}\right)$, where Ω_0 is the Rabi frequency. In contrast, first order perturbation theory is required at least to account for the high-order harmonic generation that occurs in the strong field regime of driven two-level systems [7]. In this respect, several approaches have been developed, primarily aimed at obtaining a perturbative first-order solution for the equation of motion governing the evolution of the induced dipole moment [8–10]. All these approaches have to deal with secular terms appearing at first order. These terms become divergent in the long time limit and, consequently, have to be carefully resummed in order for the perturbative solution to be applicable at any time.

A somewhat different approach, specifically designed for the strong field limit, has been followed in Ref. [11], where a first-order perturbative solution was obtained by using a series expansion dual to the Dyson series, and by resumming the corresponding secular terms by renormalization group methods [12].

Since localization can occur in a two-level system even when the strong field condition is not satisfied, it is interesting to have a perturbative solution applicable regardless of the coupling strength value. Such a solution would permit to treat within a unified formulation the localization and harmonic generation properties exhibited by this kind of systems. On the other hand, and because of the extensive use of two-level systems as a first approximation to treat more complex physical systems, analytical solutions of this type become of particular interest.

In this paper we derive a general first-order analytical solution for the perturbative evolution of a driven two-level system in the far off-resonance regime. Specifically, a first-order expression for the time-dependent density operator of the system, which is applicable regardless of the coupling strength value, is obtained. Remarkably, in the strong field limit, our perturbative expansion turns out to be valid even away from the far off-resonance condition, and therefore, in this particular case, it becomes indistinguishable from that of Ref. [11]. Moreover, our zeroth-order Hamiltonian already includes all the slowly varying contributions thus preventing, unlike previous approaches, secular terms from appearing in the corresponding first-order results.

In Sec. II we develop the formalism and obtain the dynamical evolution of the corresponding density operator. Then, in Secs. III and IV this formulation is applied, respectively, to the study of localization and high-order harmonic generation. We find that, in the absence of dissipation, driven two-level systems exhibit coherent population trapping in the far off-resonance regime. In the particular case of a symmetric double-well potential such a property implies the well-known localization of the system in one of the two wells. Finally, the main conclusions are summarized in Sec. V.

II. TIME-DEPENDENT DENSITY OPERATOR

We consider a two-level system driven by a linearly polarized laser field of frequency ω_{L} and amplitude $\mathbf{E}_0$. The energy difference between the upper level state $|2\rangle$ and the lower level state $|1\rangle$ is denoted Δ_0. In the dipole approximation the Hamiltonian reads (in atomic units, $\hbar = 1$)

$$H_d = \frac{\Delta_0}{2}\left(\sigma_{22} - \sigma_{11}\right) - \Omega_0\, g\left(t\right)\cos\left(\omega_{\mathrm{L}} t\right)\left(\sigma_{12} + \sigma_{21}\right), \tag{1}$$

where $\sigma_{ij} \equiv |i\rangle\langle j|$ is the transition operator and $\Omega_0 \equiv E_0\mu$ is the Rabi frequency, with μ being the (real) dipole matrix element between $|1\rangle$ and $|2\rangle$. It has been assumed that the laser polarization vector and the dipole moment are oriented along the same direction. Moreover, we have included in Eq. (1) an envelope function $g\left(t\right)$ in order to account for slow variations of the intensity, as occurs at the turn on and off of the laser pulse.

Appropriate scaling of the time-dependent Schrödinger equation permits the identification of the relevant dimensionless parameters. In this respect, especially suitable for our purposes is the variable change $\tau = \omega_{\mathrm{L}} t$, which yields the following dimensionless Hamiltonian

$$H = \frac{\Delta_0}{2\omega_{\mathrm{L}}}\left(\sigma_{22} - \sigma_{11}\right) - \frac{\Omega_0}{\omega_{\mathrm{L}}} f\left(\tau\right)\cos\left(\tau\right)\left(\sigma_{12} + \sigma_{21}\right), \tag{2}$$

where $f\left(\tau\right) \equiv g(\tau/\omega_{\mathrm{L}})$.

On the other hand, particularly convenient for studying the localization properties associated with the above Hamiltonian are the following coherent superposition states

$$|\mathrm{r}\rangle = \frac{1}{\sqrt{2}}\left(|1\rangle + |2\rangle\right), \tag{3}$$

$$|\mathrm{l}\rangle = \frac{1}{\sqrt{2}}\left(|1\rangle - |2\rangle\right), \tag{4}$$

which, in the case of a symmetric double-well potential, correspond to states localized on the right and left wells, respectively. In the basis $\{|\mathrm{r}\rangle, |\mathrm{l}\rangle\}$ the Hamiltonian (2) clearly displays the symmetry of the system. Indeed, in this basis Eq. (2) takes the form

$$H = -\frac{\Delta_0}{2\omega_{\mathrm{L}}}\left(\sigma_{\mathrm{lr}} + \sigma_{\mathrm{rl}}\right) - \frac{\Omega_0}{\omega_{\mathrm{L}}} f\left(\tau\right)\cos\left(\tau\right)\left(\sigma_{\mathrm{rr}} - \sigma_{\mathrm{ll}}\right), \tag{5}$$

which, for a driving field with constant amplitude ($f\left(\tau\right) = 1$), is manifestly symmetric under the combined transformation $\mathrm{r} \longleftrightarrow \mathrm{l}$ and $\tau \rightarrow \tau + \pi$.

As already said in the Introduction, in the far off-resonance regime, i.e., when $\Delta_0/\omega_{\mathrm{L}} \ll 1$, localization occurs (for $f\left(\tau\right) = 1$) at the zeros of the Bessel function $J_0\left(2\Omega_0/\omega_{\mathrm{L}}\right)$. This is the regime in which we will be primarily interested. Thus, $\Delta_0/\omega_{\mathrm{L}}$ is going to be, in principle, the small parameter of our perturbative analysis, which,

consequently, will be applicable regardless of the value of the coupling strength $\Omega_0/\omega_{\rm L}$. That is, in the far off-resonance regime our perturbative series will remain valid in both the strong and the weak field limit.

In order to obtain a small Hamiltonian, proportional to $\Delta_0/\omega_{\rm L}$, we perform the following unitary transformation

$$U(\tau) = e^{-i\phi(\tau)(\sigma_{\rm rr}-\sigma_{\rm ll})}, \tag{6}$$

with

$$\phi(\tau) = \int d\tau \frac{\Omega_0}{\omega_{\rm L}} f\left(\tau\right) \cos\left(\tau\right) \simeq \frac{\Omega_0}{\omega_{\rm L}} f\left(\tau\right) \sin\left(\tau\right), \tag{7}$$

where, in the last step, we have used the fact that the pulse envelope $f\left(\tau\right)$ is a very slowly varying function within a laser period. The transformed Hamiltonian now takes the form

$$H' = UHU^{+} - iU\dot{U}^{+} = -\frac{\Delta_0}{2\omega_{\rm L}} \left(e^{2i\phi(\tau)}\sigma_{\rm lr} + e^{-2i\phi(\tau)}\sigma_{\rm rl} \right), \tag{8}$$

where $\dot{U}^{+}$ denotes the derivative of the adjoint of U with respect to the dimensionless time τ.

In order to prevent secular terms from appearing at first order in the perturbative expansion, it is necessary to incorporate all the slowly varying terms in the zeroth-order Hamiltonian. With this purpose, and using again the fact that $f\left(\tau\right)$ hardly changes in a laser period, we express the time-dependent coefficients in Eq. (8) as the Fourier series

$$e^{\pm 2i\phi(\tau)} = \sum_{n=-\infty}^{+\infty} J_n \left[\pm \frac{2\Omega_0}{\omega_{\rm L}} f\left(\tau\right) \right] e^{in\tau} \equiv \Lambda_0 + \Lambda_{\pm}(\tau), \tag{9}$$

where

$$\Lambda_0 \equiv J_0 \left[\frac{2\Omega_0}{\omega_{\rm L}} f\left(\tau\right) \right], \tag{10}$$

$$\Lambda_{\pm}(\tau) \equiv \sum_{n=1}^{+\infty} (\pm 1)^n J_n \left[\frac{2\Omega_0}{\omega_{\rm L}} f\left(\tau\right) \right] \left(e^{in\tau} + (-1)^n e^{-in\tau} \right). \tag{11}$$

Substituting Eq. (9) into Eq. (8) we then arrive at

$$H' = H_0' + \Delta H', \tag{12}$$

with

$$H_0' \equiv -\frac{\Delta_0}{2\omega_{\rm L}} \Lambda_0 \left(\sigma_{\rm lr} + \sigma_{\rm rl} \right), \tag{13}$$

$$\Delta H' \equiv -\frac{\Delta_0}{2\omega_{\mathrm{L}}}\left(\Lambda_+(\tau)\sigma_{\mathrm{lr}} + \Lambda_-(\tau)\sigma_{\mathrm{rl}}\right). \tag{14}$$

The slowly varying part H_0' is going to be considered our zeroth-order Hamiltonian while, in the far off-resonance regime, $\Delta H'$ becomes a small perturbation. Incidentally, note that in the strong field limit, that is, when

$$\zeta \equiv \frac{2\Omega_0}{\omega_{\mathrm{L}}} f(\tau) \gg 1, \qquad \text{for } \textit{any } \tau \tag{15}$$

the Bessel functions entering Eq. (11) become, for $n \lesssim n_{\mathrm{c}} \sim \zeta$ [13],

$$J_n(\zeta) \approx \sqrt{\frac{2}{\pi\zeta}} \cos\left(\zeta - \frac{n\pi}{2} - \frac{\pi}{4}\right), \tag{16}$$

while, for $n \gtrsim n_{\mathrm{c}}$ they decay very fast as $J_n(\zeta) \sim (e\zeta/2n)^n / \sqrt{2\pi n}$. Hence, the perturbation $\Delta H'$ takes now the form

$$\Delta H' = -\frac{\Delta_0}{2\sqrt{\omega_{\mathrm{L}}\Omega_0 f(\tau)}}\left(\Pi_+(\tau)\sigma_{\mathrm{lr}} + \Pi_-(\tau)\sigma_{\mathrm{rl}}\right), \tag{17}$$

with

$$\Pi_\pm(\tau) \approx (1/\sqrt{\pi}) \sum_{n=1}^{n_{\mathrm{c}}} (\pm 1)^n \left(e^{in\tau} + (-1)^n e^{-in\tau}\right) \cos\left(\zeta - \frac{n\pi}{2} - \frac{\pi}{4}\right). \tag{18}$$

Thus, in the strong field limit, $\Delta H'$ turns out to be proportional to the parameter $\Delta_0/\sqrt{\omega_{\mathrm{L}}\Omega_0 f(\tau)}$, which has the interesting consequence that whenever the latter becomes small (for *any* τ) our perturbative results remain valid irrespective of the value of $\Delta_0/\omega_{\mathrm{L}}$.

We now proceed to solve perturbatively the quantum Liouville equation for the transformed density operator ρ', which contains all the dynamical information about the system,

$$\frac{\partial \rho'}{\partial \tau} = -i\left[H', \rho'\right] = \mathcal{L}'\rho' = \left(\mathcal{L}_0' + \Delta\mathcal{L}'\right)\rho', \tag{19}$$

$$\rho'(\tau) = U(\tau)\rho(\tau)U^+(\tau). \tag{20}$$

The linear operators $\mathcal{L}_0'$ and $\Delta\mathcal{L}'$ entering Eq. (19) represent the quantum Liouville operators corresponding to the Hamiltonians H_0' and $\Delta H'$, respectively. This evolution equation can be exactly solved to the lowest order. In doing so, one finds the following expressions for the matrix elements of the density operator in the states $|1\rangle$ and $|2\rangle$,

$$\rho_{ii}^{(0)\prime}(\tau) = \rho_{ii}'(0), \qquad i = 1, 2 \tag{21}$$

$$\rho_{12}^{(0)\prime}(\tau) = \rho'_{12}(0)e^{i\frac{\Delta_0\Lambda_0}{\omega_{\rm L}}\tau}. \tag{22}$$

It is worth noting that according to Eqs. (20) and (21), to the lowest order, the populations of the states $U^{+}(\tau)|1\rangle$ and $U^{+}(\tau)|2\rangle$ remain constant. This is a direct consequence of the fact that, for $f(\tau) \approx$ cte, the states

$$U^{+}(\tau)|1\rangle = \cos\phi(\tau)|1\rangle + i\sin\phi(\tau)|2\rangle, \tag{23}$$

$$U^{+}(\tau)|2\rangle = \sin\phi(\tau)|1\rangle - i\cos\phi(\tau)|2\rangle, \tag{24}$$

become the zeroth-order Floquet states of the system corresponding to the quasienergies $-\Delta_0\Lambda_0/2$ and $+\Delta_0\Lambda_0/2$, respectively.

The zeroth-order density operator $\rho^{(0)\prime}$ can be written as

$$\rho^{(0)\prime}(\tau) = e^{\mathcal{L}'_0\tau}\rho'(0) = \sum_{i=1,2}\rho'_{ii}(0)|i\rangle\langle i| + \left(\rho'_{12}(0)e^{i\frac{\Delta_0\Lambda_0}{\omega_{\rm L}}\tau}|1\rangle\langle 2| + \text{h.c.}\right). \tag{25}$$

Notice that this expression provides, in turn, an useful operational definition for the evolution operator $e^{\mathcal{L}'_0\tau}$.

With the aim of deriving a first-order perturbative expression for the time-dependent density operator, we rewrite the quantum Liouville equation (19) in the interaction representation with respect to $\mathcal{L}'_0$. Specifically,

$$\frac{\partial\rho'_{\rm I}}{\partial\tau} = \Delta\mathcal{L}'_{\rm I}\,\rho'_{\rm I}(\tau), \tag{26}$$

where

$$\rho'_{\rm I} = e^{-\mathcal{L}'_0\tau}\rho'(\tau), \tag{27}$$

$$\Delta\mathcal{L}'_{\rm I} = e^{-\mathcal{L}'_0\tau}\Delta\mathcal{L}'e^{\mathcal{L}'_0\tau}. \tag{28}$$

The solution of Eq. (26) can be expressed as the following infinite series

$$\rho'_{\rm I}(\tau) = \rho'_{\rm I}(0) + \int_0^\tau d\tau_1\Delta\mathcal{L}'_{\rm I}(\tau_1)\rho'_{\rm I}(0) + \int_0^\tau d\tau_1\int_0^{\tau_1} d\tau_2\Delta\mathcal{L}'_{\rm I}(\tau_1)\Delta\mathcal{L}'_{\rm I}(\tau_2)\rho'_{\rm I}(0) + \ldots \tag{29}$$

This is a perturbative expansion in the small parameter $\Delta_0/\omega_{\rm L}$ which, in the strong field limit, becomes an expansion in the small parameter $\Delta_0/\sqrt{\omega_{\rm L}\Omega_0 f(\tau)}$, as already said. Thus, up to first order, one obtains

$$\rho'(\tau) = e^{\mathcal{L}'_0\tau}\rho'(0) + e^{\mathcal{L}'_0\tau}\int_0^\tau d\tau_1 e^{-\mathcal{L}'_0\tau_1}\Delta\mathcal{L}'(\tau_1)e^{\mathcal{L}'_0\tau_1}\rho'(0) + O\left(\epsilon^2\right), \tag{30}$$

where ϵ represents, in general, the small parameter characterizing the expansion. On the other hand, the integrand on the right hand side of Eq. (30) takes the form

$$
e^{\mathcal{L}_0'(\tau-\tau_1)}\Delta\mathcal{L}'(\tau_1)e^{\mathcal{L}_0'\tau_1}\rho'(0) = -ie^{\mathcal{L}_0'(\tau-\tau_1)}\left[\Delta H'(\tau_1), e^{\mathcal{L}_0'\tau_1}\rho'(0)\right]
$$

$$
= \sum_{i=1,2}\xi_{ii}'(\tau_1)|i\rangle\langle i| + \left(\xi_{12}'(\tau_1)e^{i\frac{\Delta_0\Lambda_0}{\omega_\mathrm{L}}(\tau-\tau_1)}|1\rangle\langle 2| + \mathrm{h.c.}\right), \tag{31}
$$

where

$$
\xi_{jk}'(\tau) \equiv -i\langle j|\left[\Delta H'(\tau), e^{\mathcal{L}_0'\tau}\rho'(0)\right]|k\rangle, \tag{32}
$$

and the definition of the propagator $e^{\mathcal{L}_0'(\tau-\tau_1)}$, as provided by Eq. (25), has been used. Substituting then Eq. (25) into Eq. (32) one obtains, after some lengthy algebra,

$$
\xi_{jj}'(\tau) \equiv (-1)^j\frac{\Delta_0}{\omega_\mathrm{L}}\sum_{n=0}^{+\infty}J_{2n+1}(\zeta)\sin[(2n+1)\tau]\left(\rho_{12}'(0)e^{i\frac{\Delta_0\Lambda_0}{\omega_\mathrm{L}}\tau} + \mathrm{h.c.}\right), \tag{33}
$$

$$
\begin{aligned}
\xi_{12}'(\tau) \;\equiv\; & -\frac{\Delta_0}{\omega_\mathrm{L}}\sum_{n=0}^{+\infty}J_{2n+1}(\zeta)\sin[(2n+1)\tau]\left[\rho_{22}'(0)-\rho_{11}'(0)\right] \\
& + 2i\frac{\Delta_0}{\omega_\mathrm{L}}\sum_{n=1}^{+\infty}J_{2n}(\zeta)\cos(2n\tau)\,\rho_{12}'(0)e^{i\frac{\Delta_0\Lambda_0}{\omega_\mathrm{L}}\tau},
\end{aligned} \tag{34}
$$

$$
\xi_{21}'(\tau) = \xi_{12}'^*(\tau), \tag{35}
$$

with $\zeta = 2\Omega_0 f(\tau)/\omega_\mathrm{L}$. Substitution of the above expressions into Eq. (31) and then into Eq. (30) leads to the following final result

$$
\begin{aligned}
\rho'(\tau) = & \left[\rho_{11}'(0) + \frac{\Delta_0}{\omega_\mathrm{L}}\alpha(\tau)\right]|1\rangle\langle 1| + \left[\rho_{22}'(0) - \frac{\Delta_0}{\omega_\mathrm{L}}\alpha(\tau)\right]|2\rangle\langle 2| \\
& + \left\{\left[\rho_{12}'(0) + \frac{\Delta_0}{\omega_\mathrm{L}}\beta(\tau)\right]e^{i\frac{\Delta_0\Lambda_0}{\omega_\mathrm{L}}\tau}|1\rangle\langle 2| + \mathrm{h.c.}\right\} + O\left(\epsilon^2\right),
\end{aligned} \tag{36}
$$

where

$$
\alpha(\tau) \equiv \frac{1}{2}\sum_{n=0}^{+\infty}\frac{J_{2n+1}(\zeta)}{2n+1}\left\{\rho_{12}'(0)\left(e^{i\left[(2n+1)+\frac{\Delta_0\Lambda_0}{\omega_\mathrm{L}}\right]\tau} + e^{-i\left[(2n+1)-\frac{\Delta_0\Lambda_0}{\omega_\mathrm{L}}\right]\tau} - 2\right) + \mathrm{h.c.}\right\}, \tag{37}
$$

$$
\begin{aligned}
\beta(\tau) \;\equiv\; & \frac{1}{2}\sum_{n=0}^{+\infty}\frac{J_{2n+1}(\zeta)}{2n+1}\left[\rho_{22}'(0)-\rho_{11}'(0)\right]\left\{e^{i\left[(2n+1)-\frac{\Delta_0\Lambda_0}{\omega_\mathrm{L}}\right]\tau} + e^{-i\left[(2n+1)+\frac{\Delta_0\Lambda_0}{\omega_\mathrm{L}}\right]\tau} - 2\right\} \\
& + 2i\sum_{n=1}^{+\infty}\frac{J_{2n}(\zeta)}{2n}\rho_{12}'(0)\sin(2n\tau).
\end{aligned} \tag{38}
$$

Thus, the first-order density operator of the driven two-level system is finally given by

$$\rho(\tau) = U^{+}(\tau)\rho'(\tau)U(\tau), \tag{39}$$

with $U(\tau)$ defined in Eqs. (6) and (7). This expression, which contains no secular terms, is our central result and is applicable not only in the far off-resonance limit, i.e.,

$$\Delta_0/\omega_{\rm L} \ll 1, \tag{40}$$

but also in the regime where

$$\Delta_0/\omega_{\rm L} \ll \sqrt{\Omega_0 f\,(\tau)\,/\omega_{\rm L}} \gg 1. \tag{41}$$

III. COHERENT POPULATION TRAPPING AND LOCALIZATION

Coherent population trapping and localization properties are conveniently analyzed in terms of the diagonal matrix elements of $\rho(\tau)$ in the basis $\{|\mathrm{r}\rangle, |\mathrm{l}\rangle\}$. By using Eqs. (3) and (39) one can express the population $\langle \mathrm{r}|\rho(\tau)|\mathrm{r}\rangle$ as

$$\rho_{\rm rr}(\tau) = \frac{1}{2}\left\{1 + \left[\rho'_{12}(\tau) + \rho'_{21}(\tau)\right]\right\}. \tag{42}$$

Taking into account that $\rho'(0) = \rho(0)$ one obtains, after substitution of Eq. (22) into Eq. (42), the following zeroth-order expression

$$\rho^{(0)}_{\rm rr}(\tau) = \frac{1}{2} + |\rho_{12}(0)| \cos\left(\frac{\Delta_0\Lambda_0}{\omega_{\rm L}}\tau + \varphi\right), \tag{43}$$

where $\rho_{12}(0) = |\rho_{12}(0)|e^{i\varphi}$. Equation (43) shows that, in the absence of dissipation, the populations of the $|\mathrm{r}\rangle$ and $|\mathrm{l}\rangle$ states, which in general oscillate with a frequency $\Delta_0\Lambda_0$ (curves (a) and (b) in Fig. 1), become trapped whenever the parameters of the driving field are so tuned that the Bessel function $\Lambda_0 \equiv J_0\left(2\Omega_0 f\,(\tau)\,/\omega_{\rm L}\right)$ attains a zero (curve (c) in Fig. 1). Remarkably, such a population trapping occurs regardless of the system initial state. This is a general zeroth-order result that remains valid as long as conditions (40) or (41) are satisfied. In the particular case of a symmetric double-well potential, $|\mathrm{r}\rangle$ and $|\mathrm{l}\rangle$ become states localized on the right and left wells, respectively, so that, when the system is initially in either of the two wells, one obtains the peculiar localization property reported in previous works [2–6].

Substituting Eq. (36) into Eq. (42) one finds the following final expression for the first-order time-dependent population

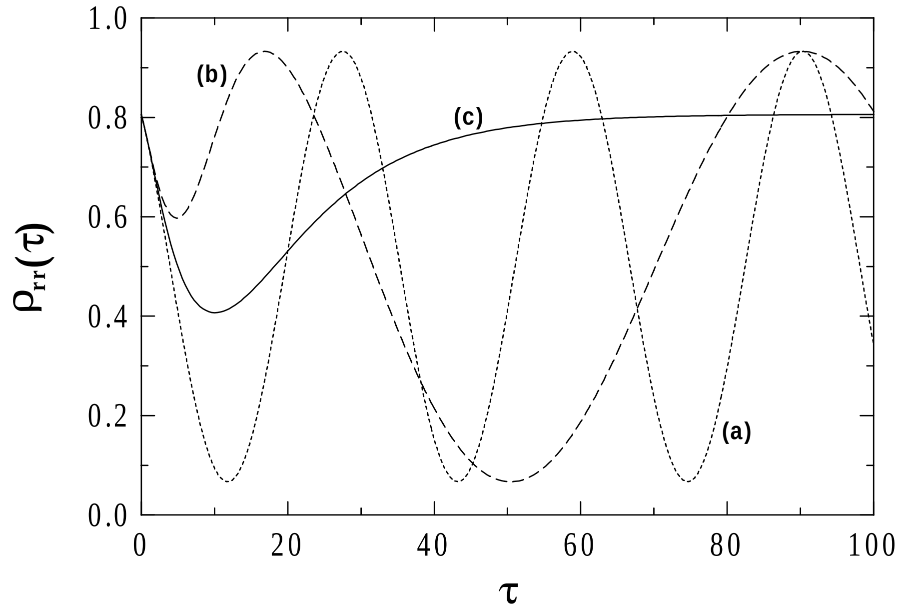

Figure 1: Zeroth-order population $\rho_{\rm rr}(\tau)$ for a pulse envelope $f(\tau) = 1 - \exp(-\tau/10)$ and $\Delta_0/\omega_{\rm L} = 0.2$, $|\rho_{12}(0)| = \sqrt{3}/4$, $\varphi = \pi/4$; (a) $\Omega_0/\omega_{\rm L} = 0$; (b) $\Omega_0/\omega_{\rm L} = 1.8$; (c) $\Omega_0/\omega_{\rm L} = 1.202$.

$$\begin{aligned}\rho_{\rm rr}(\tau) \;&=\; \frac{1}{2} + |\rho_{12}(0)| \cos\left(\frac{\Delta_0\Lambda_0}{\omega_{\rm L}}\tau + \varphi\right) \\ &\quad + \frac{\Delta_0}{\omega_{\rm L}}\left[\rho_{22}(0) - \rho_{11}(0)\right] \sum_{n=0}^{+\infty} \frac{J_{2n+1}(\zeta)}{2n+1} \left(\cos(2n+1)\tau - \cos\frac{\Delta_0\Lambda_0}{\omega_{\rm L}}\tau\right) \\ &\quad - 2\frac{\Delta_0}{\omega_{\rm L}}|\rho_{12}(0)| \sum_{n=1}^{+\infty} \frac{J_{2n}(\zeta)}{2n} \sin(2n\tau) \sin\left(\frac{\Delta_0\Lambda_0}{\omega_{\rm L}}\tau + \varphi\right) + O\left(\epsilon^2\right). \qquad (44)\end{aligned}$$

Equation (44) shows that now the populations evolve undergoing rapidly oscillating changes of the order of ϵ, superimposed to the much slower dominant oscillating evolution of frequency $\Delta_0\Lambda_0$ (see curve (a) in Fig. 2). In general, these high frequency oscillations cannot be eliminated by the external field and as a consequence, unlike the previous case, to this order it is not possible to achieve exact coherent trapping for any system initial state. Indeed, when $\Lambda_0 = 0$ the population $\rho_{\rm rr}(\tau)$ evolves describing small-amplitude rapid oscillations (with frequencies that are integer multiples of the laser frequency) about the initial population $\rho_{\rm rr}(0)$ [curve (b) in Fig. 2]. Yet, exact first-order coherent trapping can still be obtained by properly choosing the initial state. Taking $\rho_{11}(0) = \rho_{22}(0) = 1/2$ and $\varphi = 0$, Eq. (44) becomes, when $\Lambda_0 = 0$,

$$\rho_{\rm rr}(\tau) = \frac{1}{2} + \rho_{12}(0) = \rho_{\rm rr}(0). \qquad (45)$$

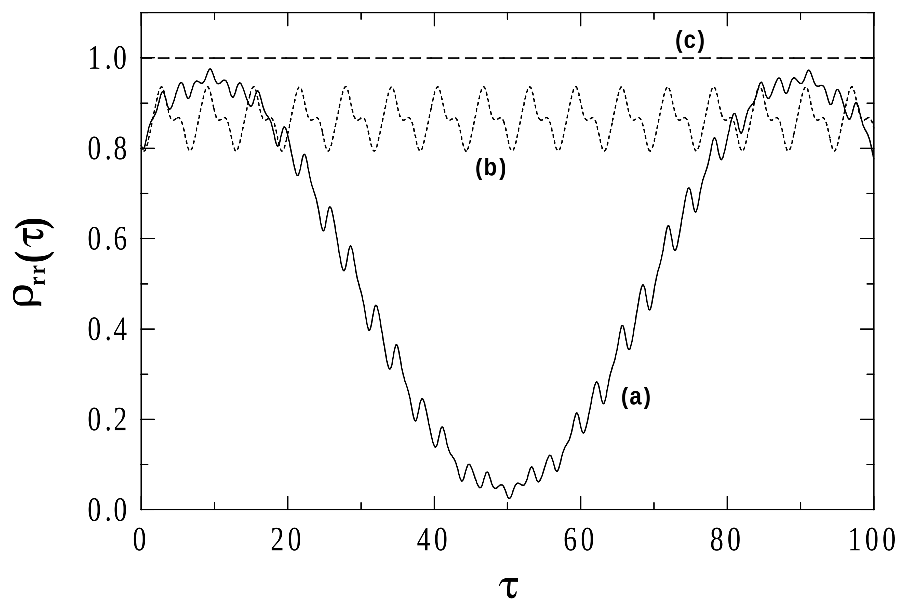

Figure 2: First-order population $\rho_{\rm rr}(\tau)$ for $f(\tau) = 1$ and $\Delta_0/\omega_{\rm L} = 0.2$; (a) $\rho_{11}(0) = 3/4$, $\rho_{22}(0) = 1/4$, $|\rho_{12}(0)| = \sqrt{3}/4$, $\varphi = \pi/4$, $\Omega_0/\omega_{\rm L} = 1.8$; (b) $\rho_{11}(0) = 3/4$, $\rho_{22}(0) = 1/4$, $|\rho_{12}(0)| = \sqrt{3}/4$, $\varphi = \pi/4$, $\Omega_0/\omega_{\rm L} = 1.202$; (c) $\rho_{11}(0) = \rho_{22}(0) = 1/2$, $|\rho_{12}(0)| = 1/2$, $\varphi = 0$, $\Omega_0/\omega_{\rm L} = 1.202$.

That is, when the system is initially in either the state $|{\rm r}\rangle$ or $|{\rm l}\rangle$, first-order coherent population trapping can be achieved by simply choosing the parameters of the driving field in such a way that $\Lambda_0 = 0$ (curve (c) in Fig. 2). Notice that, as before, in the special case of a symmetric double-well potential, this result implies the localization of the system in one of the two wells.

On the other hand, it is interesting to note that according to Eq. (44) the population of a system initially located in the right well of a symmetric double-well potential, evolves in the limit of zero driving field as

$$\rho_{\rm rr}(t) = \cos^2\left(\frac{\Delta_0}{2}t\right) \tag{46}$$

so that, the well-known result for the undriven system is recovered, as it should be.

Notice that the pulse envelope $f(t)$ simply modifies the way in which the system reaches its steady state (see Fig. 1). As a consequence, localization cannot be perturbatively achieved starting from an initial delocalized eigenstate. Indeed, as is apparent from Eq. (44), in this case the system remains always delocalized, within terms of the order of ϵ. This result, which is in contradiction with the numerical results obtained in Refs. [3,4], simply reflects that such a localization process is a nonperturbative effect that cannot be accounted for by our treatment.

IV. HARMONIC GENERATION

Driven two-level systems has been extensively used as convenient models to understand the basic mechanism underlying the phenomenon of high-order harmonic generation [7,8,10,14–16]. This is so primarily because, as pointed out by Sundaram and Milonni [7], such a simple system already exhibits the main features observed experimentally in the emission spectrum of atoms in very intense laser fields, namely, the existence of a plateau in the harmonic spectrum followed by a sharp cutoff. This, in turn, is an indication of the fact that such features are intrinsic properties of strongly driven systems.

The purpose of this Section is to show how the previous formulation leads straightforwardly to the well-known results of high-order harmonic generation in strongly driven two-level systems.

The coherent part of the emission spectrum $S(\omega)$ is proportional to $|d(\omega)|^2$, with $d(\omega)$ being the Fourier component at the frequency ω of the induced dipole moment $\langle d(t)\rangle$ [7,14]

$$S(\omega) \propto |d(\omega)|^2 = \left| \frac{1}{T} \int_{t_0}^{t_0+T} dt\, e^{i\omega t} \langle d(t)\rangle \right|^2 . \tag{47}$$

The time evolution of $\langle d(t)\rangle$ follows directly from Eq. (44). Indeed, taking into account that the quantum dipole operator is given by

$$d = \mu\,(\sigma_{12} + \sigma_{21}) = \mu\,(\sigma_{\rm rr} - \sigma_{\rm ll})\,, \tag{48}$$

one finds

$$\langle d(t)\rangle = {\rm Tr}\,[\rho(t) d] = \mu\,[\rho_{\rm rr}(t) - \rho_{\rm ll}(t)] = 2\mu\,(\rho_{\rm rr}(t) - 1/2)\,, \tag{49}$$

where, in the last step, we have used that $\rho_{\rm rr}(t) + \rho_{\rm ll}(t) = 1$. Therefore, by substituting Eq. (44) into Eq. (49), and then this latter into Eq. (47) one can immediately obtain the corresponding emission spectrum. In doing so, one finds the well-known result that $S(\omega)$ consists, in general, of three types of peaks:

i) Low frequency components located at $\omega = \pm\Delta_0\Lambda_0$ and intensities proportional to

$$|d_{\pm\Delta_0\Lambda_0}|^2 = \mu^2 |\rho_{12}(0)|^2. \tag{50}$$

ii) Hyper-Raman lines located at frequencies $2n\omega_{\rm L} \pm \Delta_0\Lambda_0$ and intensities proportional to

$$|d_{2n\omega_{\rm L}\pm\Delta_0\Lambda_0}|^2 = \left| \mu \frac{\Delta_0}{\omega_{\rm L}} |\rho_{12}(0)| \frac{J_{2n}(\zeta)}{2n} \right|^2 \qquad n = 1, 2, 3, \ldots \tag{51}$$

iii) Odd harmonic components located at frequencies $(2n+1)\,\omega_{\rm L}$ and intensities proportional to

$$\left|d_{(2n+1)\omega_{\mathrm{L}}}\right|^2 = \left|\mu\frac{\Delta_0}{\omega_{\mathrm{L}}}\left[\rho_{22}(0) - \rho_{11}(0)\right]\frac{J_{2n+1}(\zeta)}{2n+1}\right|^2 \qquad n = 0, 1, 2, \ldots \tag{52}$$

Formulas of this type have been previously obtained by Ivanov and Corkum [8] and Dakhnovskii and Bavli [10].

It is interesting to note that exact first-order coherent population trapping (localization) has a twofold manifestation in the system spectroscopic properties: a zero frequency peak appears and the spectrum consists of only even harmonic components.

On the other hand, the appearance in the strong field regime ($\zeta \gg 1$) of the plateau and corresponding cutoff in the harmonic spectrum follows from the asymptotic behaviour of the Bessel functions, which for $n \lesssim n_{\mathrm{c}} \sim \zeta$ are given by Eq. (16) while for $n \gtrsim n_{\mathrm{c}}$ they drop very rapidly as $(e\zeta/2n)^n / \sqrt{2\pi n}$.

V. CONCLUSION

Driven two-level systems display a number of interesting features that are common to more general driven systems. This fact, makes them convenient starting points for analyzing more complex physical systems. Moreover, under certain circumstances, they allow for an analytical treatment which would be otherwise impossible. Such analytical treatments provide detailed information on the system response to specific changes in the external parameters, and this information can be used to control its dynamical evolution.

In this work we have studied analytically the time evolution of driven two-level systems in the far off-resonance regime ($\Delta_0/\omega_{\mathrm{L}} \ll 1$). This was done by performing a perturbative analysis based on a convenient zeroth-order Hamiltonian which takes care of divergent secular terms. In this way, we obtained a general first-order expression for the time-dependent density operator which is valid regardless of the coupling strength value. Interestingly enough, in the strong field regime, this expression turns out to be applicable even when the far off-resonance condition is not satisfied. Indeed, the perturbative analysis remains valid as long as the condition $\Delta_0/\omega_{\mathrm{L}} \ll \sqrt{\Omega_0 f(\tau)/\omega_{\mathrm{L}}} \gg 1$ holds.

The analytical formulation presented in this paper makes it possible to treat in a unified framework different aspects of the dynamical evolution of driven two-level systems. In particular, from the time evolution of the populations it immediately follows that the well-known phenomenon of tunneling suppression in a symmetric double-well potential can be considered as a specific manifestation of a more general population trapping phenomenon.

To the lowest order, regardless of the system initial state, the populations of the coherent superpositions $|\mathrm{r}\rangle$ and $|\mathrm{l}\rangle$ become trapped, in the absence of dissipation, whenever the parameters of the driving field are tuned in such a way that the Bessel function

$J_0 \left(2\Omega_0 f\left(\tau\right)/\omega_{\mathrm{L}}\right)$ vanishes. To first-order, however, small-amplitude rapid oscillations in the time evolution of the populations appear, which, in general, cannot be eliminated by the external field. As a consequence, to this order, such a result only remains valid in an approximate way. Nonetheless, exact first-order coherent population trapping still occurs when the system is initially in either the state $|\mathrm{r}\rangle$ or $|\mathrm{l}\rangle$. In the particular case of a symmetric double-well potential, in which $|\mathrm{r}\rangle$ and $|\mathrm{l}\rangle$ become states localized on the right and left wells, respectively, this result implies the localization of the system in one of the two wells.

The present formulation also leads straightforwardly to the well-known results of high-order harmonic generation in strongly driven two-level systems.

Of course physical systems exhibit a number of interesting features which are of nonperturbative origin and, consequently, cannot be accounted for by a perturbative treatment of the type presented here. In spite of this, the present formulation can be useful as a starting point for analyzing a large variety of driven systems in the perturbative regime.

ACKNOWLEDGMENTS

This work has been supported by DGESIC (Spain) under Project No. PB97-1479-C02-01.

REFERENCES

1. For a review see, for example, M. Grifoni and P. Hänggi, Phys. Rep. **304**, 229 (1998).
2. F. Grossmann, T. Dittrich, P. Jung, and P. Hänggi, Phys. Rev. Lett. **67**, 516 (1991).
3. R. Bavli and H. Metiu, Phys. Rev. Lett. **69**, 1986 (1992).
4. R. Bavli and H. Metiu, Phys. Rev. A **47**, 3299 (1993).
5. J. M. Gomez Llorente and J. Plata, Phys. Rev. A **45**, R6958 (1992).
6. F. Grossmann and P. Hänggi, Europhys. Lett. **18**, 571 (1992).
7. B. Sundaram and P. W. Milonni, Phys. Rev. A **41**, R6571 (1990).
8. M. Yu. Ivanov and P. B. Corkum, Phys. Rev. A **48**, 580 (1993).
9. Y. Dakhnovskii and H. Metiu, Phys. Rev. A **48**, 2342 (1993).
10. Y. Dakhnovskii and R. Bavli, Phys. Rev. B **48**, 11020 (1993).
11. M. Frasca, Phys. Rev. A **60**, 573 (1999).
12. M. Frasca, Phys. Rev. A **56**, 1548 (1997).
13. M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1972).
14. F. I. Gauthey, C. H. Keitel, P. L. Knight, and A. Maquet, Phys. Rev. A **52**, 525 (1995).
15. M. Pons, R. Taïeb, and A. Maquet, Phys. Rev. A **54**, 3634 (1996).
16. F. I. Gauthey, C. H. Keitel, P. L. Knight, and A. Maquet, Phys. Rev. A **55**, 615 (1997).